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Donor and acceptor concentrations in degenerate InN

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A formalism is presented to determine donor (N_D) and acceptor (N_A) concentrations in wurtzitic InN characterized by degenerate carrier concentration (n) and mobility (μ). The theory includes scattering not only by charged point defects and impurities, but also by charged threading dislocations, of concentration N_{dis} . For a 0.45- μm -thick InN layer grown on Al_2O_3 by molecular beam epitaxy, having $N_{\text{dis}} = 5 \times 10^{10} \text{ cm}^{-2}$, determined by transmission electron microscopy, $n(20 \text{ K}) = 3.5 \times 10^{18} \text{ cm}^{-3}$ and $\mu(20 \text{ K}) = 1055 \text{ cm}^2/\text{Vs}$, determined by Hall effect measurements, the fitted values are $N_D = 4.7 \times 10^{18} \text{ cm}^{-3}$ and $N_A = 1.2 \times 10^{18} \text{ cm}^{-3}$. The identities of the donors and acceptors are not known, although a comparison of N_D with analytical data, and also with calculations of defect formation energies, suggests that a potential candidate for the dominant donor is H. © 2002 American Institute of Physics. [DOI: 10.1063/1.1432742]

Interest in III-N materials has mushroomed in recent years because of new developments in blue/UV light-emitting diodes (LEDs) and laser diodes (LDs), and also high-frequency transistors operating at high powers and temperatures.¹ The LEDs, LDs, and transistors typically involve GaN, InGaN, and AlGaIn, with low In and Al fractions in the latter two cases, respectively. However, there would be great advantages in being able to incorporate large fractions of In and Al in various applications. For example, the use of wurtzitic InN would permit photonic devices in the red (band gap $\sim 1.89 \text{ eV}$), and much faster electronic devices, because of higher mobility and peak velocity than those of most other III-N materials. Indeed, a room-temperature mobility $\mu = 2700 \text{ cm}^2/\text{Vs}$, for a carrier concentration $n = 5 \times 10^{16} \text{ cm}^{-3}$, was measured many years ago in a sample grown by rf reactive ion sputtering (rf-RIS).² However, there have been no recent reports of such high mobilities or low carrier concentrations, and present activity seems to be concentrated on applying some of the more popular growth techniques, such as molecular beam epitaxy (MBE),^{3–6} and metalorganic chemical vapor deposition (MOCVD).⁷ So far, the highest room-temperature mobility measured in our MBE-grown InN is $1310 \text{ cm}^2/\text{Vs}$, with the highest reported MOCVD mobility somewhat lower.⁷ The problem seems to be the extremely high concentrations of donors incorporated, $> 10^{18} \text{ cm}^{-3}$. These donors have not yet been identified, but potential candidates are defects, such as the N vacancy, and impurities, such as O_N , Si_{In} , and possibly interstitial H.

From theoretical calculations, Stampfl *et al.*⁸ find that O_N and Si_{In} have lower formation energies than those of any of the native donor-type defects in n -type InN.

In this work, we will deal with state-of-the-art MBE growth of InN on Al_2O_3 . To make further progress in MBE and MOCVD InN, and to approach the rf-RIS results,² it will be necessary to identify and eliminate the major source of donors. We have earlier developed a simple general model⁹ to determine donor (N_D) and acceptor (N_A) concentrations in degenerate semiconductor material, such as InN/ Al_2O_3 , with high concentrations of impurities, point defects, and dislocations. From a comparison with glow discharge mass spectroscopy (GDMS) measurements, and the theory mentioned above, it appears that the high values of N_D could possibly be explained by H impurity, but not by O or Si. However, native defects also cannot be completely ruled out.

The growth of InN was performed with a turbopumped Varian GEN-II gas-source MBE chamber with a background pressure of 1×10^{-10} Torr, conventional effusion cells, and an EPI unibulb rf plasma source for generation of nitrogen radicals.^{3,5} The nitrogen flux through the plasma source was fixed at 0.4 sccm, causing a nitrogen partial pressure in the MBE chamber of 1.0×10^{-5} Torr during growth. The growth process began with nitridation of (0001) Al_2O_3 wafers at a temperature of 400°C for 30 min. Next, a 0.2- μm -thick AlN nucleation layer was deposited at 1050°C , and finally, a 0.45- μm -thick InN film, at 460°C . Both AlN and InN growths were performed by the conventional MBE technique.

The mismatches in lattice constant and thermal expansion coefficient between InN and AlN are expected to generate high densities of threading edge, screw, and mixed dislocations. For this particular material, transmission electron microscopy (TEM) measurements, shown in Fig. 1, find about $3 \times 10^{10} \text{ cm}^{-2}$ edge dislocations, and $2 \times 10^{10} \text{ cm}^{-2}$

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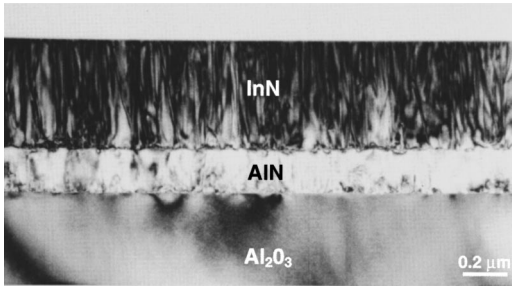


FIG. 1. Cross-sectional transmission electron microscope image of a MBE-grown InN layer.

screw and mixed dislocations. The total concentration of threading dislocations in the InN layer is about four times higher than that in the AlN buffer layer, which is unexpected since the AlN/Al₂O₃ lattice mismatch is not much different than the InN/AlN mismatch ($\sim 12\% - 13\%$). Thus, other factors, such as interface impurities, may generate further dislocations. Oxygen would be a candidate here because of its high affinity for Al; however, GDMS measurements, discussed below, do not find significant quantities of O in the bulk of the InN layer.

In *n*-type GaN, it is known that threading edge dislocations are negatively charged,¹⁰ and, under certain conditions, the same is true of screw and mixed dislocations.¹¹ The reason for such charging is fundamental, i.e., the formation energy of a negatively charged, shallow-acceptor species, such as a Ga vacancy along the dislocation core, is lowered by about the band-gap energy, and thus makes such an acceptor species much more probable in *n*-type material.¹² Now, InN has a lower band gap than that of GaN, only 1.9 eV compared to 3.5 eV, but nevertheless, the same considerations still apply. Thus, for purposes of this study, we will assume that all threading dislocations are negatively charged, with about 1 *e* per *c*-lattice distance ($c = 5.71$ Å). However, we will also show that this assumption does not have a strong effect on the value of N_D determined from our formalism.

The van der Pauw–Hall effect measurements were performed with a LakeShore Model 7507 apparatus, including a closed-cycle He cooling system operating from 15 to 320 K. From measurements of Hall coefficient R and conductivity σ , the Hall mobility $\mu_H = R\sigma$ and the Hall concentration $n_H = 1/eR$ could be calculated at each temperature. These results are shown in Fig. 2. It is seen that μ_H and n_H vary little as a function of temperature, and thus may be considered degenerate. In that case, $\mu_H = \mu_c$, the conductivity mobility, and $n_H = n$, the true carrier concentration.¹³ The existence of degeneracy in InN, with $n = 3.5 \times 10^{18} \text{ cm}^{-3}$, would also have been predicted by the calculated concentration at the Mott transition, about $4 \times 10^{16} \text{ cm}^{-3}$.

Scattering theory for degenerate electrons at low temperatures is straightforward because: (1) n is simply related to the Hall coefficient R in degenerate material, i.e., $n = 1/eR$; (2) phonon scattering is negligible at low T , and the only important mechanisms are dislocation scattering and ionized point-defect/impurity scattering; (3) the degeneracy of the electrons allows the application of Matthiessen's rule: $\mu_{\text{tot}}^{-1} = \mu_{\text{dis}}^{-1} + \mu_{\text{ion}}^{-1}$; and (4) the relaxation-time approximation holds for these two elastic scattering mechanisms.^{9,13} Earlier we showed that⁹

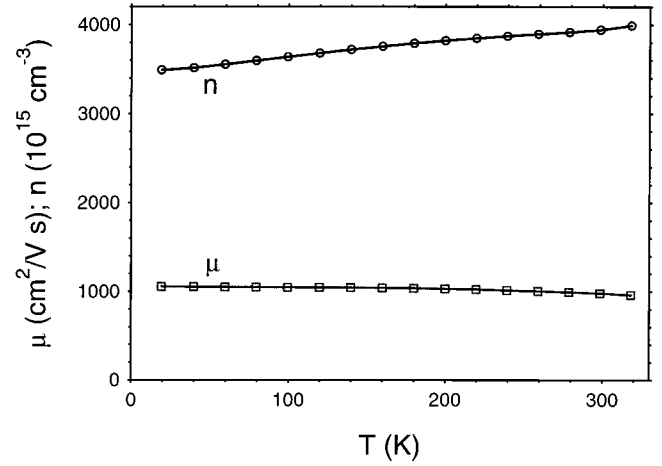


FIG. 2. Temperature dependence of mobility and carrier concentration for a MBE-grown InN layer.

$$\mu_{\text{dis,deg}} = \frac{e \tau_{\text{dis}}(E_F)}{m^*} = \frac{4 \times 3^{2/3} e c^2 n^{2/3}}{\pi^{8/3} \hbar N_{\text{dis}}} [1 + y(n)]^{3/2}, \quad (1)$$

where N_{dis} is the dislocation density, c is the *c*-lattice constant, and

$$y(n) = \frac{2 \cdot 3^{1/3} \pi^{8/3} \hbar^2 \epsilon n^{1/3}}{e^2 m^*}. \quad (2)$$

Also, it is well known that

$$\mu_{\text{ion,deg}} = \frac{24 \pi^3 \epsilon^2 \hbar^3 n}{e^3 m^{*2} N_{\text{ion}} \left[\ln[1 + y(n)] - \frac{y(n)}{1 + y(n)} \right]} \quad (3)$$

where N_{ion} is the density of ionized impurities and point defects. Equation (3) is, of course, the degenerate form of the familiar Brooks–Herring formula.¹³ For $n > 10^{18} \text{ cm}^{-3}$, we can assume that an impurity band is formed, and that all donors are ionized, as in a metal. Then, charge balance dictates that $N_D = N_A + n + N_{\text{dis}}/c$, where we have explicitly included the negatively charged dislocations as part of the charge balance. Furthermore, $N_{\text{ion}} = N_D + N_A = 2N_A + n + N_{\text{dis}}/c$, so that if N_{dis} is known, the only unknown in Eqs. (1)–(3) is N_A . The combined mobility is now simply given by Matthiessen's rule, since the electrons are degenerate (energy independent):

$$\mu_{\text{tot}}^{-1} = \mu_{\text{dis,deg}}^{-1} + \mu_{\text{ion,deg}}^{-1}. \quad (4)$$

Note that Eq. (4) is a transcendental equation in only one variable, N_A , since μ_{tot} and n are determined from the Hall effect measurements, and N_{dis} from the TEM measurements. Note further that, remarkably, the only physical constants required in this formalism are the effective mass m^* ($= 0.11 m_0$),¹⁴ the static dielectric constant ϵ ($= 15.3 \epsilon_0$),¹⁵ and the *c*-lattice constant ($= 5.71$ Å).¹⁵ The situation is much more complicated in nondegenerate material at higher temperatures.¹³

The sample shown in Figs. 1 and 2 has $\mu = 1055 \text{ cm}^2/\text{V s}$, $n = 3.49 \times 10^{18} \text{ cm}^{-3}$, at 20 K, and $N_{\text{dis}} = 5 \times 10^{10} \text{ cm}^{-2}$. Inserting these numbers into Eq. (4) gives $N_A = 7.5 \times 10^{17}$ and $N_D = 5.1 \times 10^{18} \text{ cm}^{-3}$. If we assume that only the edge dislocations are charged, then $N_{\text{dis}} = 3 \times 10^{10} \text{ cm}^{-2}$, giving $N_A = 1.4 \times 10^{18} \text{ cm}^{-3}$, and $N_D = 5.4$

$\times 10^{18} \text{ cm}^{-3}$. If we assume that no dislocations are charged, then $N_A = 2.4 \times 10^{18} \text{ cm}^{-3}$ and $N_D = 5.9 \times 10^{18} \text{ cm}^{-3}$. Thus, we can get a fairly accurate value for N_D , no matter what is assumed about N_{dis} . Analyses of other samples, from different growth runs, give similar results.

To compare N_D with the concentrations of various donor-type impurities, we have obtained GDMS analysis¹⁶ for six selected elements, with the following results: $[\text{C}] = 1 \times 10^{18}$, $[\text{O}] = 3 \times 10^{16}$, $[\text{Al}] = 2 \times 10^{16}$, $[\text{Si}] = 2 \times 10^{16}$, $[\text{S}] = 4 \times 10^{15}$, and $[\text{H}] = 9 \times 10^{19} \text{ cm}^{-3}$. These numbers may be considered accurate within a factor two.¹⁶ Noting that $N_D = 5 \times 10^{18} \text{ cm}^{-3}$, we can state that, among these elements, only H could be the dominant donor. In particular, neither O nor Si qualify, even though, from theoretical calculations,⁸ they both have low formation energies. Coupled with the high formation energies for the native donor defects,⁸ the most likely candidate for the dominant donor, in this particular InN material, is H. However, this assignment must be considered very tentative.

In summary, we have presented a formalism to determine donor N_D and acceptor N_A concentrations in degenerate semiconductor films. For a 0.45- μm -thick InN film grown by MBE, with mobility $\mu = 1055 \text{ cm}^2/\text{V s}$ and carrier concentration $n = 3.49 \times 10^{18} \text{ cm}^{-3}$, at 20 K, and threading dislocation density $N_{\text{dis}} = 5 \times 10^{10} \text{ cm}^{-2}$, we have calculated $N_A = 7.5 \times 10^{17} \text{ cm}^{-3}$ and $N_D = 5.1 \times 10^{18} \text{ cm}^{-3}$. A tentative candidate for the dominant donor is H.

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